Understanding the Nature of Marine Aerosols and Their Effects in the Coupled Ocean-Atmosphere System

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LONG-TERM GOALS

The long-term goal of this work is to understand the nature of marine aerosols and how these particles influence visibility, cloud microphysics and precipitation, the thermodynamic structure of the marine boundary layer, and the transmission of radiation.

OBJECTIVES

The objectives of this project extend across three areas: (i) advancing aerosol measurement techniques via the development of a new instrument to quantify aerosol-water interactions, (ii) improving knowledge and model predictions related to the physicochemical nature of aerosols and ocean-aerosol-cloud-precipitation-radiation interactions, and (iii) strengthening a research methodology leveraging multiple complementary tools of analysis to guide future studies of this nature in the marine atmosphere.

APPROACH

The main technical approach is to use a combination of in-situ aircraft measurements, cloud models, and satellite remote sensors to study the nature and character of aerosols and their effects in the marine atmosphere over a broad range of spatial and temporal scales. This work includes the following tasks:

- Develop and characterize a state-of-the-art instrument to quantify aerosol-water interactions, which will be deployed in future aircraft studies.
- Use satellite remote sensing observations to quantify and interrelate measurements of ocean bio-optical properties, aerosol and cloud microphysics, radiative properties, and precipitation in the marine atmosphere, while accounting for meteorology.
- Use cloud models to examine the salient features of cloud drop activation and the subsequent growth of drops to precipitation-sized drops over a wide range of conditions, and the capability to more easily differentiate between aerosol and meteorological effects on cloud microphysics, precipitation, and radiative transfer.

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- Carry out in-situ measurements in the marine atmosphere to simultaneously characterize aerosol physicochemical properties and to quantify the interactive nature between aerosols, oceans, clouds, precipitation, meteorology, and radiation.

Key scientists involved with this work include Dr. Fred Brechtel (Brechtel Manufacturing Inc.), who is collaborating with the PI on the development of a new instrument to quantify aerosol hygroscopicity. Dr. Graham Feingold (National Oceanic and Atmospheric Administration) has collaborated on using large eddy simulation to study aerosol effects in the marine atmosphere. Dr. Haflidi Jonsson at the Naval Postgraduate School is instrumental in the aircraft operations and the data management, which is critical for the aircraft data analysis tasks in this project.

WORK COMPLETED

The major work completed thus far includes the continued development of a novel aerosol hygroscopic growth measurement probe, and advancing studies on aerosol-cloud-precipitation interactions in the marine atmosphere. Three peer-reviewed publications have resulted from the latter effort thus far in the project lifetime.

RESULTS

Despite decades of research devoted to aerosol-cloud interactions, the response in precipitation resulting from aerosol perturbations is highly uncertain owing to a number of macrophysical and microphysical processes that occur and interact over a wide range of temporal and spatial scales. Three separate studies discussed below have been carried out to help improve quantification of aerosol-cloud-rain interactions using models and observational data.

Study 1. Deconstructing the Precipitation Susceptibility Construct: Improving Methodology for Aerosol-Cloud-Precipitation Studies (Sorooshian et al., 2010).

In previous work the concept of precipitation susceptibility was introduced to serve as a framework for quantifying aerosol effects on precipitation (Sorooshian et al., 2009). Precipitation susceptibility is defined as

$$S_o = -\frac{d \ln R}{d \ln N_d} \tag{1}$$

where R is precipitation rate and N_d is cloud drop number concentration. The minus sign is applied so that a positive value of S_o reflects the conventional wisdom that aerosol abundance and R are negatively correlated for warm-rain clouds. Using both models and satellite observations, an analysis of shallow cumulus clouds showed that S_o is strongly controlled by cloud liquid water path (LWP) and exhibits three regimes: (i) low LWP, where clouds do not precipitate and are relatively insensitive to N_d; (ii) intermediate LWP, where precipitation is progressively more effectively suppressed by increasing N_d; and (iii) high LWP, where susceptibility begins to decrease because there is ample LWP to drive the precipitation process.

The absolute value of the precipitation susceptibility and its behavior as a function of macrophysical conditions (e.g. LWP) for individual cloud regimes are highly uncertain. Precipitation susceptibility is difficult to quantify with observations owing to the limitations of current instrumentation, the cost of dedicated field studies, and a lack of long-term datasets needed to provide statistical significance for a sufficiently large range of aerosol variability influencing specific cloud regimes over a range of macrophysical conditions. These issues motivated this study to improve the quantification of S_o using a diverse set of synergistic tools including models of varying complexity, satellite observations from NASA's A-Train, and Navy Twin Otter aircraft measurements.

An alternative definition of Eq. 1 was introduced to directly relate precipitation changes to aerosol perturbations using satellite data,

$$S_o' = -\frac{d\ln R}{d\ln\alpha}, \tag{2}$$

where N_d is replaced with a sub-cloud CCN proxy (α = aerosol index), which can be retrieved by space-borne remote sensors such as MODIS. Both forms of the susceptibility are proposed to also be quantified using the product of two separate metrics termed ACI and χ:

$$S_o = -\left.\frac{\partial\ln r_e}{\partial\ln N_d}\right|_{LWP}\cdot\chi \tag{3}$$

$$S_o' = ACI\ \cdot\chi\ , \tag{4}$$

where the two sub-components are defined as

$$\chi = \left.\frac{\partial\ln R}{\partial\ln r_e}\right|_{LWP} \tag{5}$$

$$\mathrm{ACI} = -\left.\frac{\partial\ln r_e}{\partial\ln\alpha}\right|_{LWP}. \tag{6}$$

It has been shown with basic assumptions that ACI is bounded by the range 0 – 0.33 (Feingold et al., 2001), where the maximum value corresponds to complete activation of sub-cloud aerosols

($= -\left.\frac{\partial\ln r_e}{\partial\ln N_d}\right|_{LWP}$). The hypothesis is that by separating precipitation susceptibility into two

components, the statistical significance and confidence in causal relationships between aerosol and precipitation can be increased, while also circumventing some problems associated with the interpretation of satellite data, specifically the difficulty of measuring aerosol in the vicinity of clouds. It is noted that the χ metric completely avoids the need to use collocated aerosol and cloud remote sensing data. The main findings of this work are as follows:

The response of precipitation to a change in cloud optical depth (or alternatively drop size), defined by χ, captures the essence of precipitation susceptibility behavior. The link between aerosol and cloud microphysics (ACI) has a dampening effect on the absolute value of susceptibility (in the form of S_o') as compared to χ. This is exacerbated by the tendency for ACI to be underestimated by satellite analyses.

The deconstruction of the precipitation susceptibility into components (Eqs. 3 and 4) is shown to qualitatively reproduce the LWP-dependent behavior of the directly-quantified precipitation susceptibility for shallow cumulus clouds (Eqs. 1 and 2). The absolute agreement between is remarkably close using aircraft measurements of stratocumulus clouds and based on simulations of shallow cumulus clouds using models of varying complexity. Therefore, separating the precipitation susceptibility into components provides more confidence in the microphysical chain of events leading from aerosol perturbations to changes in precipitation rate.

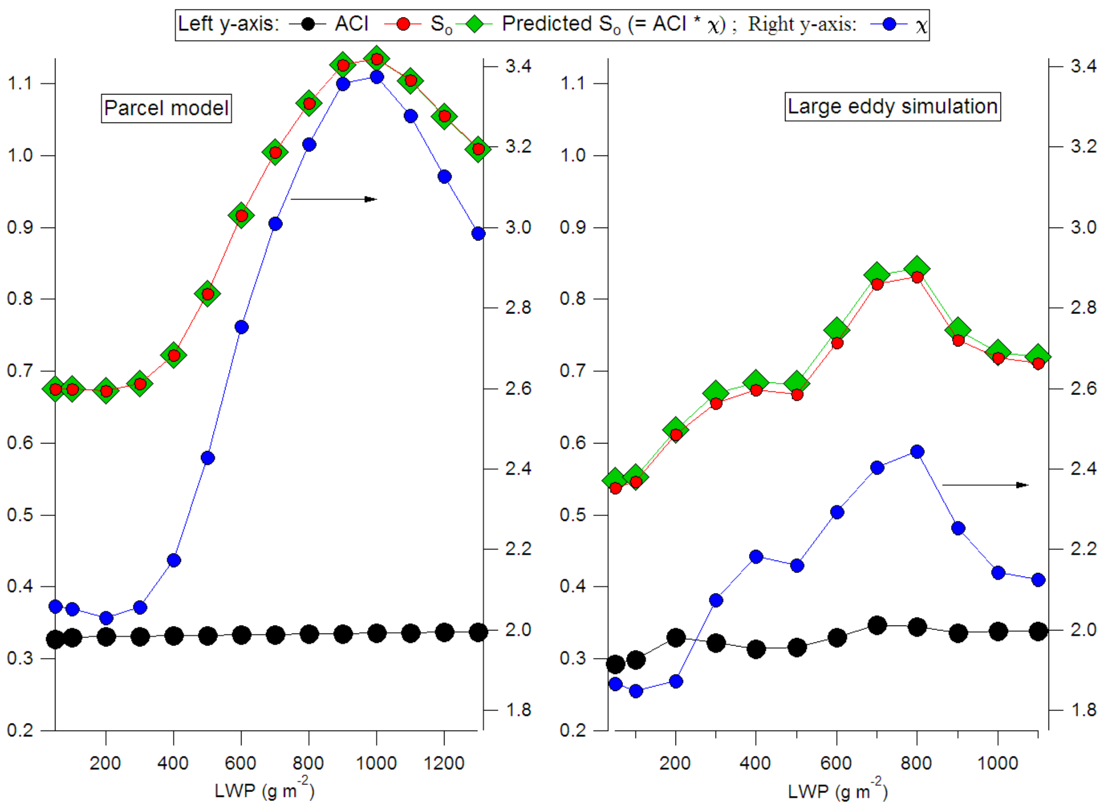

Figure 1. Predicted $-\left.\dfrac{\partial \ln r_e}{\partial \ln N_d}\right|_{LWP}$ ***(= 0.33, which is equivalent to maximum value of ACI),*** χ***, and*** S_o

as a function of cloud liquid water path (LWP) from a (**a**) parcel model and (**b**) large eddy simulation. Here it is shown that χ is clearly driving the behavior of S_o, where ACI simply has a dampening effect on the absolute value of S_o as compared to χ. The product of ACI and χ is shown to be almost identical to the directly quantified value of S_o (Eq. 1).

Study 2. Effect of aerosol on the susceptibility and efficiency of precipitation in warm trade cumulus clouds (Jiang et al., 2010)

Observations of stratocumulus clouds suggest that rain rate (R) can be expressed as a simple power-law function of liquid water path (LWP) and N_d (e.g. Pawlowska and Brenguier 2003). Whether the relationship derived from stratocumulus can be applied to shallow cumulus convection, given their different dynamical and morphological characteristics, is unknown. This motivated a study to examine the relationship between precipitation rate R, liquid water path LWP, droplet concentration N_d, and for the first time to consider the effect of cloud lifetime on R in a model-generated population of cumulus clouds. The inclusion of cloud lifetime is important in that the development of precipitation depends on the cloud microphysical environment (broadly defined by N_d and liquid water content, LWC), and also the time available for the collision-coalescence processes (Feingold et al., 1996). Populations of cumulus clouds exposed to different aerosol concentrations were modeled using large eddy simulation to explore the effect of aerosol (via its influence on drop concentration) on the precipitation, precipitation efficiency, and precipitation susceptibility of cumulus clouds. The major results are summarized as follows:

Clouds with diameters less than 500 m produce little precipitation, although they contribute significantly to cloud fraction and cloud reflectance. In moderately polluted conditions (200 cm^{-3} $\leq N_a \leq$ 300 cm^{-3}), clouds can grow as large and as deep as in clean conditions (N_a = 100 cm^{-3}), but produce less rain (Note: N_a corresponds to aerosol concentration). Although the larger/deeper clouds occur infrequently, they generate most of the precipitation. The majority of clouds have lifetimes less than 30 min. For small, short-lived clouds, lifetimes are insensitive to aerosol conditions. Considering the subset of the largest clouds, polluted clouds tend to have longer lifetimes due to precipitation reduction. One caveat is that for the most polluted conditions studied (N_a = 500 cm^{-3}), the largest clouds do not grow to be as big or to live as long as their moderately polluted counterparts.

Precipitation rate (R) can be reasonably well-represented by a power-law function of LWP and N_d. The respective powers for LWP and N_d are of similar magnitude to those of observational and LES studies of stratocumulus clouds, despite the differences in characteristics and morphology of the two types of clouds. The time-integrated precipitation rate is well-represented by a power-law function of LWP, N_d, and cloud lifetime.

Analysis of S_o with respect to controlling parameters other than LWP, such as the ratio of autoconversion to accretion rates, yields a similar result (Fig. 2) since the ratio of autoconversion to accretion rates is strongly correlated to LWP. Also, a similar result is obtained for S_o versus the ratio of the time scales for condensation relative to that for rain production (τ_{cond}/τ_{driz}). This is because a cloud in its formative stages will have small LWP and therefore small τ_{cond}/τ_{driz}, owing to the microphysics being dominated by condensation and τ_{driz} being relatively large compared to τ_{cond}.

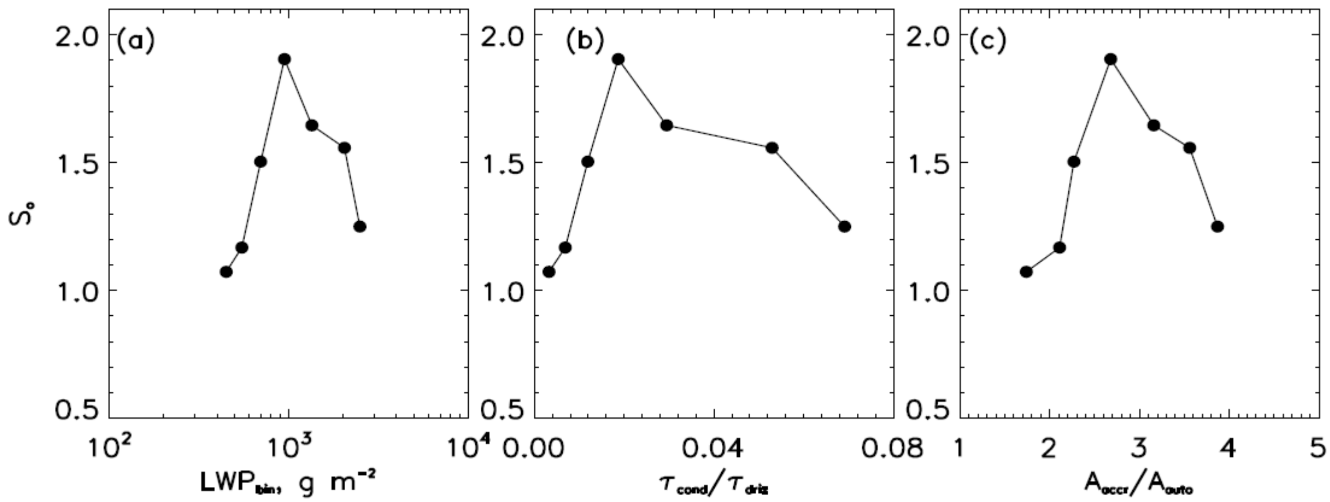

Figure 2. (a) Precipitation susceptibility (S_o) vs. LWP, (b) S_o vs. τ_{cond}/τ_{driz}, and (c) S_o vs. A_{accr}/A_{auto}. A_{auto} and A_{accr} are the rates of autoconversion and accretion, respectively. τ_{cond} is the microphysical condensation time scale, τ_{driz} (= $r_l/(A_{accr}+A_{auto})$) represents a time scale for rain production, and r_l denotes liquid water mixing ratio.

Study 3. Ocean Emission Effects on Aerosol-Cloud Interactions: Insights from Two Case Studies (Sorooshian and Duong, 2010)

Two case studies were conducted to evaluate the effect of ocean emissions on aerosol-cloud interactions. A review of the first case study from the eastern Pacific Ocean showed that simultaneous aircraft and space-borne observations are valuable in detecting links between ocean biota emissions and marine aerosols. For example, increasing chlorophyll A corresponded to enhancements in particulate concentrations of diethylamine and methanesulfonate, both of which originate from ocean-derived biogenic sources. But the effect of the ocean biota emissions on cloud microphysics is shown to be less clear owing to interference from background anthropogenic pollution and the difficulty with field experiments in obtaining a wide range of aerosol conditions to robustly quantify ocean effects on aerosol-cloud interactions. To address these limitations, the second case study leveraged remote sensing data over the less polluted Southern Ocean region. The results indicate that cloud drop size is reduced more for a fixed increase in aerosol particles during periods of higher ocean chlorophyll A (Figure 3). This can potentially be explained by there being aerosols with more favorable physicochemical properties for droplet activation during periods of higher chlorophyll A. This has important implications for marine boundary layer microphysics, as these aerosols are more hygroscopic and can scatter more light and may be more effective cloud condensation nuclei.

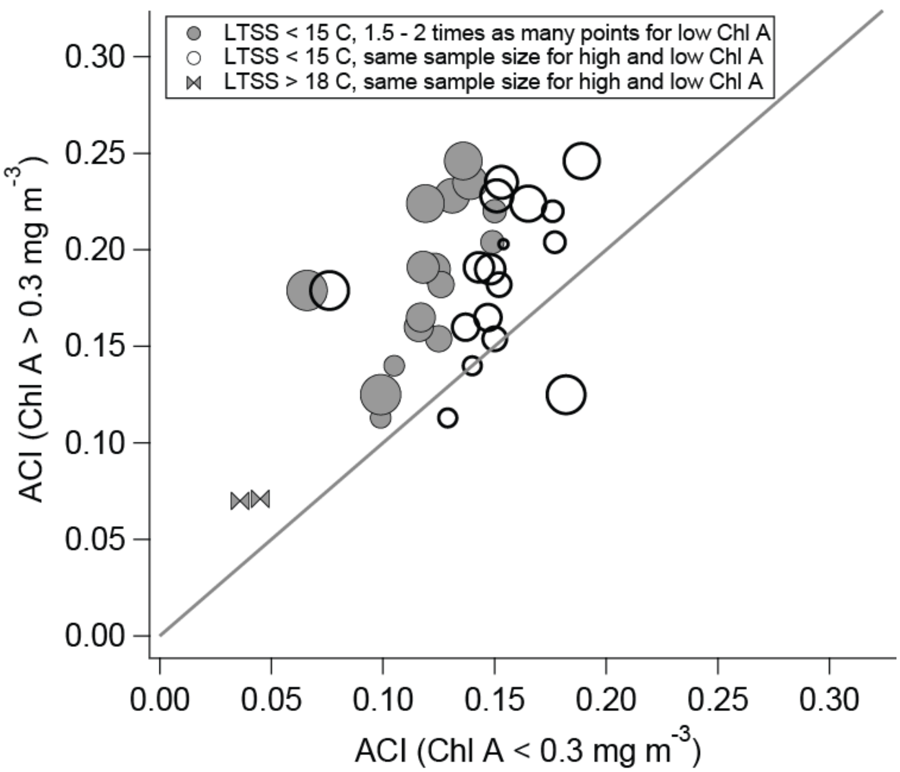

Figure 3. Comparison of ACI values ($ACI = -\left.\frac{\partial \ln r_e}{\partial \ln \alpha}\right|_{LWP}$ ***) for periods of relatively high and low chlorophyll A concentration in the Southern Ocean region based on data over a span of 27 months starting in June 2006. The solid line represents the 1:1 line. ACI is a proxy for Aerosol-Cloud Interactions, r_e is cloud drop effective radius, and α is a measure of aerosol abundance, which in this case is aerosol index as retrieved by MODIS. Marker sizes are proportional to cloud liquid water path (LWP) between 50 and 350 g m^{-2} (11 total LWP bins). LTSS corresponds to lower tropospheric static stability, where values below 15^o C correspond to relatively unstable atmospheric conditions and values exceeding 18^o C represent stable conditions.***

IMPACT/APPLICATIONS

The method to study aerosol-cloud-rain interactions outlined in Sorooshian et al. (2010) is expected to be of important application in future studies, especially as the amount of data being collected by satellite remote sensors is growing. An issue in future studies will be the difficulty of relating aerosol perturbations to cloud microphysics and especially precipitation, and the deconstruction of the precipitation susceptibility is specifically meant to help address some of these issues.

The knowledge gained from the Jiang et al. (2010) study can be tested against field observations or remote sensing data, as in Sorooshian et al. (2010). Various parameters derived from the relationship between precipitation, and microphysical variables may be useful in parameterizing cloud and precipitation processes in large scale models. For example, probability distribution functions of LWP, N_d, and cloud lifetime derived from the simulations presented in this study for weakly precipitating trade cumulus could be used to represent the probability distribution of *R* for the trade cumulus cloud regime without explicitly relying on poorly-resolved convection and cloud processes at the large-scale.

RELATED PROJECTS

There are no related projects at this time.

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PUBLICATIONS

Sorooshian, A., G. Feingold, M. D. Lebsock, H. Jiang, and G. Stephens (2010), Deconstructing the precipitation susceptibility construct: Improving methodology for aerosol-cloud-precipitation studies, *J. Geophys. Res.*, *115*, D17201, doi:10.1029/2009JD013426. [published, refereed]

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HONORS/AWARDS/PRIZES

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Sorooshian, A., G. Feingold, M. D. Lebsock, H. Jiang, and G. Stephens (2010), Deconstructing the precipitation susceptibility construct: Improving methodology for aerosol-cloud-precipitation studies, *J. Geophys. Res*., *115*, D17201, doi:10.1029/2009JD013426.

Invitee to 8th Annual NCAR Early Career Scientist Assembly Junior Faculty Forum (2010) (Armin Sorooshian, University of Arizona)